

ontario hydro research division

COMPLIMENTARY COPY

Please Credit Ontario Hydro Research if
Reproduced in Whole or in Part

To: Mr. R.G. Lalonde
Technical Superintendent
Darlington Operations

HT DEPOSITION TO DARLINGTON SOILS

F.S. Spencer

Soil samples collected from 8 representative sites at Darlington NGS were exposed to the HT plume during the 10 June, 1987 Chalk River HT field release experiment. HT deposition velocities were 1.4×10^{-4} to $11.6 \times 10^{-4} \text{ m s}^{-1}$, in good agreement with those measured during both the 1986 and 1987 Chalk River HT field experiments and various laboratory and field chamber experiments. These results suggest that the rate of HT plume depletion at Darlington by soil oxidation during temperatures supporting biological activity would be similar to results observed at Chalk River and other locations given a normal range of soil water content and free pore volume.

INTRODUCTION

The objective of this work was to determine under field conditions the potential of various soils of the Darlington site to oxidize molecular tritium, HT, to tritiated water, HTO, a much more radiotoxic form of tritium. With HT as a major atmospheric source term at the Darlington Tritium Recovery Facility knowledge of HT oxidation is necessary for reliable tritium environmental modeling and dose estimation. Focus was placed on soils because laboratory and field studies have shown that the dominant mechanism for oxidation of an HT plume is soil microbial conversion.

METHODS

Soil was sampled from representative areas of the Darlington property, Figure 1, based on vegetation cover and expected soil type. The sampling sites included two cultivated areas, (1 and 5), two deciduous woodlots, (2 and 6), two hydroseeded areas, (3 and 4), and two old fields (7 and 8).

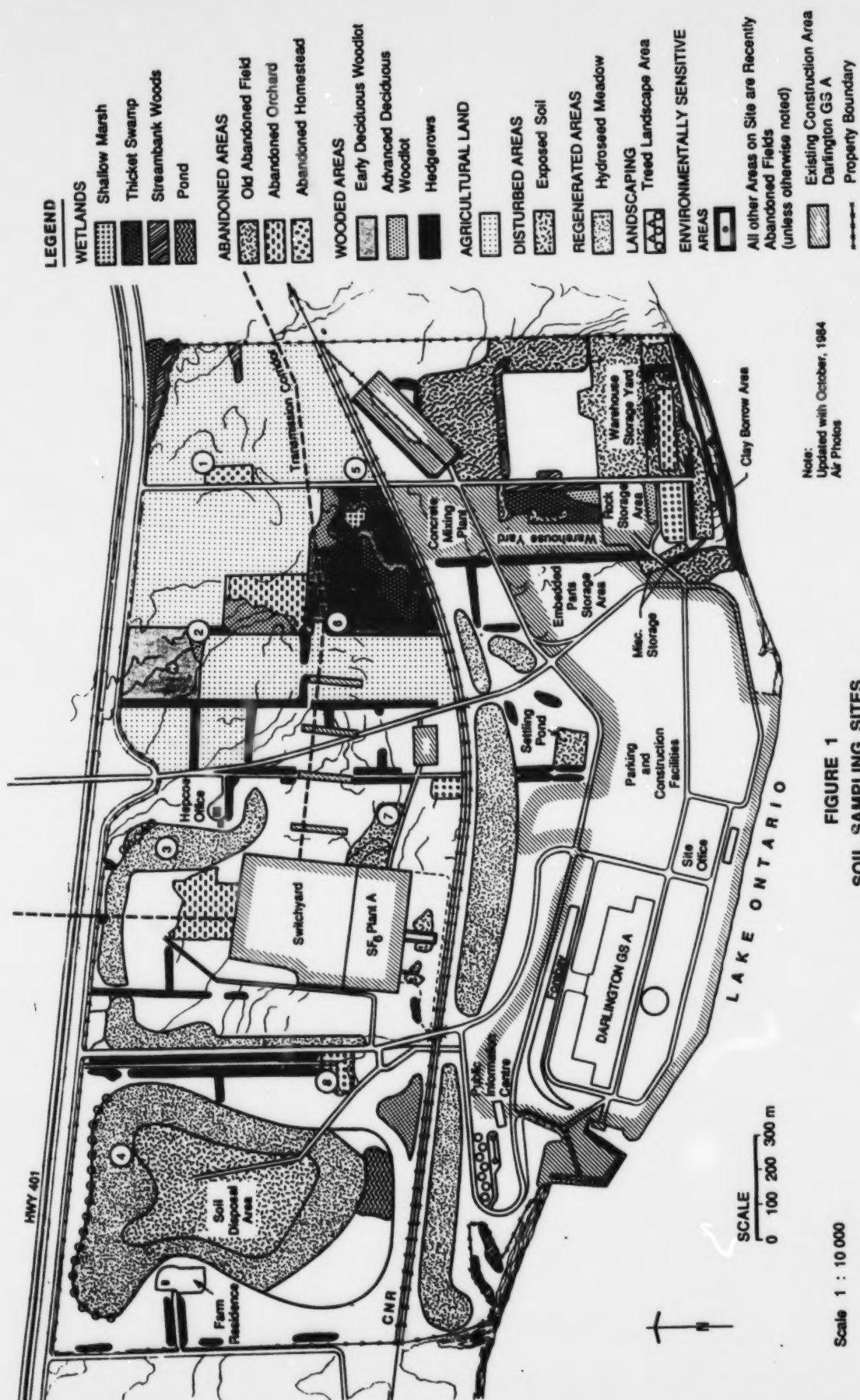
Four soil cores of 6.3 cm diameter and 22 cm depth were taken at each site. Heavy-walled PVC tubes with a removable stainless steel lip were used for sampling. After sampling, the lower ends of sampling tubes were sealed with polyisobutylene tape (water impermeable) and the upper ends covered with cheesecloth to maintain aerobic conditions. Ambient soil moisture and microbial activity were maintained by transporting the samples to Chalk River in a humidified cooler.

job	34-167-402-50	file	837.88	date	September 12, 1988	report	88-198-K
-----	---------------	------	--------	------	--------------------	--------	----------

LEGAL NOTICE

This report was prepared by Ontario Hydro and, while effort has been made to assure the accuracy and applicability of the material contained herein, neither Ontario Hydro nor any person acting on its behalf:

- (a) makes any warranty, express or implied with respect to the use of any information, apparatus, method or process disclosed in this report or that such use may not infringe privately owned rights; or
- (b) assumes any liabilities with respect to the use of, or for damages resulting from the use of, any information, apparatus, method or process disclosed in this report.



Three cores of each site were exposed to the HT plume during the 10 June, 1987 field release experiment at Chalk River. Exposure took place 51 m downwind from the release site (see Figure 1 in CFFTP Report No CFFTP-G-88007 by Brown *et al* 1988). Soil cores were left intact in sampling tubes and the upper surface at 22 cm height exposed to the HT plume. Soil cores were exposed to HT 13 days after Darlington sample collection.

The tritium source term was 3.54 TBq HT released over 30 minutes at 1 m height on the upwind side of an open field. The atmospheric concentration 50 m downwind was determined by sampling with palladium impregnated Molecular Sieve 4A traps. From 5 to 30 minutes during the release the time-integrated air concentration was $1.455 \times 10^6 \text{ Bq m}^{-3}$.

Within 30 minutes after exposure cores were sealed with Parafilm to prevent moisture (and HTO) loss. Soil was removed from most sampling tubes 3 to 4 hours after exposure and stored in glass jars for transport in coolers to Research Division. HTO was extracted by azeotropic distillation and counted by liquid scintillation spectrometry using Monophase 40 Plus (see Methods in CFFTP Report No CFFTP-G-88007 by Brown *et al* 1988).

Water content and bulk density (dry wt/soil volume) of soil cores exposed to HT (3 per site) were determined by oven drying at 105°C. Soil porosity was calculated using measured bulk density and an assumed particle density of 2.65 g cm^{-3} (Brady 1974): $\% \text{ porosity} = 100 - (\text{bulk density/particle density} \times 100)$. Soil free pore volume (volume of soil pores not filled with water) was calculated as follows: $\% \text{ free pore volume} = \% \text{ porosity} - (\% \text{ water content by volume})$ the latter calculated from water content in $\text{g H}_2\text{O/g dry wt}$, bulk density in g cm^{-3} . Soil parameters are shown in Table 1.

The fourth core of each site was submitted to the University of Guelph Soil Testing Laboratory for soil texture, organic content and pH measurements.

RESULTS AND DISCUSSION

The uptake of HT by soil or other substrates is most often characterized by a deposition velocity, defined as the ratio of deposition flux to the atmospheric concentration of HT. Time averaged deposition velocities were calculated from the HTO accumulated in each soil core (soil deposit, Appendix 1) divided by the 25 minute time-integrated HT concentration in air 50 m downwind from the release point. This HT air concentration at 1 m height was $1.455 \times 10^6 \text{ Bq m}^{-3}$ from 5 to 30 minutes during the release (Brown *et al* 1988). Calculation of a rate parameter such as deposition velocity allows comparison of values obtained from various experiments.

The mean deposition velocities ranged from a low of $1.4 \times 10^{-4} \text{ m s}^{-1}$ at site 7 to $11.6 \times 10^{-4} \text{ m s}^{-1}$ at site 4, Figure 2. The variability within sites expressed as the coefficient of variation (SD % of mean) was generally less than 50% which is normal for these type of measurements. There appeared to be some dependence of mean deposition velocity on type of vegetation cover with one hydroseeded site, number 4, having the highest value and an old field site, number 7, the lowest value. This may reflect the expected relative fertility of these sites. Corn sites 1 and 5 plus deciduous sites 2 and 6 were about the same (5×10^{-4} to $7 \times 10^{-4} \text{ m s}^{-1}$) while large differences occurred between hydroseeded sites 3 and 4 and between old field sites 7 and 8. Slightly higher deposition velocities were observed in the forest area than the open field at Chalk River during the 1987 HT field release experiment (Table 10 in Brown *et al* 1987). Studies of German soils by Förstel (1988) demonstrated forest soil deposition velocities about 5 to 10 times those of field soils.

TABLE 1

SOIL PROPERTIES

Soil particle is shown as a per cent of total particle weight classified as follows: sand 53 to 200 μm , silt 2 to 53 μm , and clay less than 2 μm diameter. Organic and water content are expressed on a dry weight basis. A single core was used for soil texture (sand, silt, clay), organic content and pH. Triplicate cores were used for all other values.

	Soil Type	Soil Composition			Organic Content (% of (dry wt)	pH	Water Content ($\bar{x}$, % of dry wt)	Bulk Density ($\bar{x}$, g cm^{-3})	Porosity ($\bar{x}$, %)	Free Pore Volume ($\bar{x}$, %)
		Sand (%)	Silt (%)	Clay (%)						
1	Clay loam	26.5	46.1	27.4	5.8	6.4	22.9	1.14	57.1	31.1
2	Loam	40.6	41.6	17.8	6.9	6.9	39.5	1.01	61.8	21.2
3	Loam	33.6	49.3	17.0	0.9	7.4	42.0	1.07	59.6	14.8
4	Loam	43.9	43.5	12.6	4.6	7.3	13.5	1.66	37.4	14.9
5	Fine Sandy Loam	52.8	32.8	14.5	4.3	6.9	27.3	1.14	56.8	25.6
6	Loam	28.9	47.0	24.1	5.3	7.0	28.2	1.23	53.4	18.7
7	Loam	37.2	42.4	20.4	2.5	7.0	22.1	1.48	44.2	11.6
8	Fine Sandy Loam	53.5	30.6	15.9	1.7	7.2	22.4	1.35	49.0	18.8

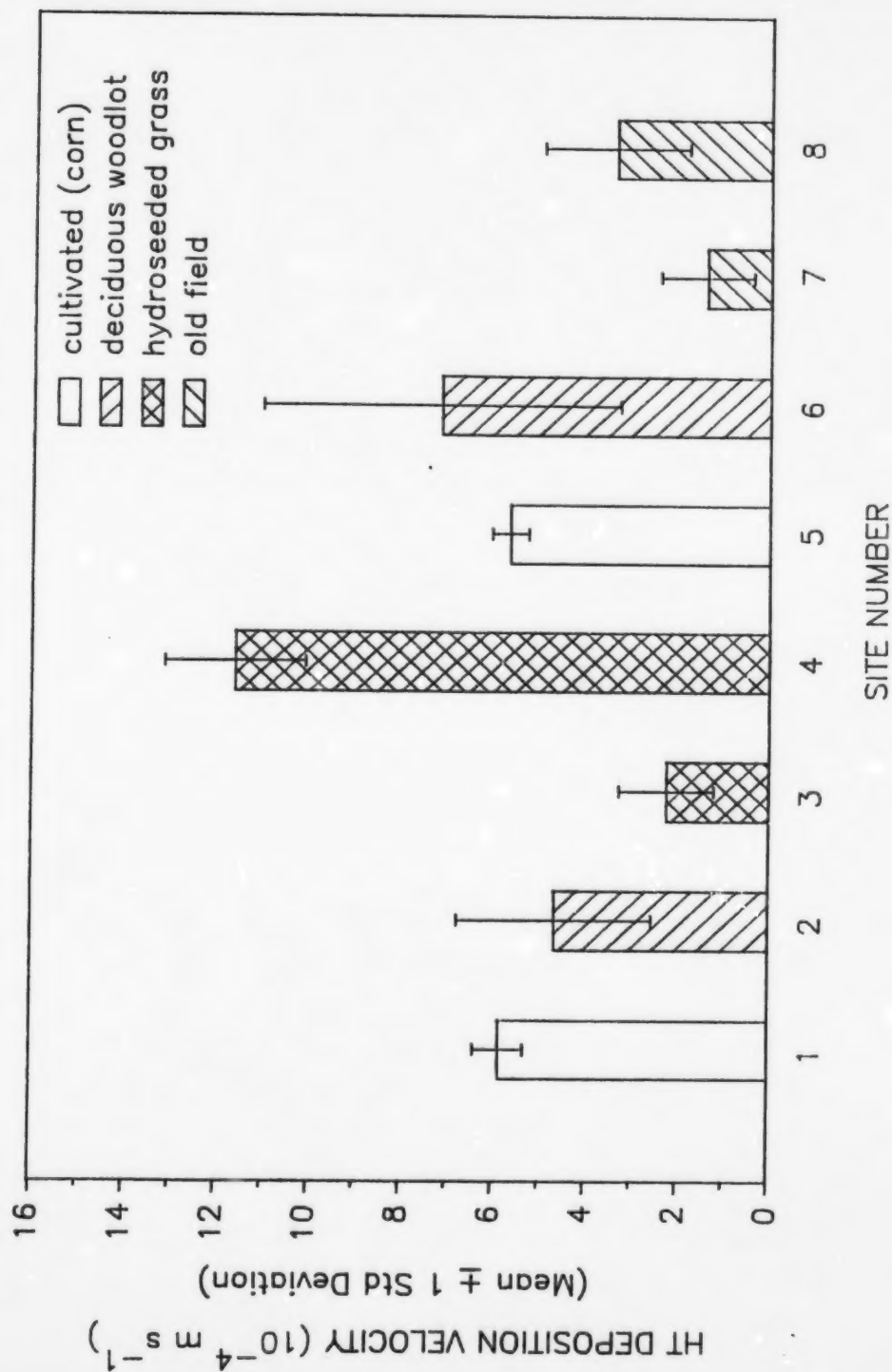


Figure 2. Darlington Soil HT Deposition Velocities

The Darlington soil HT deposition velocities agreed well with those measured at Chalk River during the same HT release. At the same exposure location (43 to 50 m downwind) Darlington values of 1.4×10^{-4} to $11.6 \times 10^{-4} \text{ m s}^{-1}$ compare to Chalk River values of 4×10^{-4} to $11 \times 10^{-4} \text{ m s}^{-1}$, (see Table 10 in CFFTP Report No CFFTP-G-88007 by Brown *et al* 1988). The range of Chalk River deposition velocities extends to 2.7×10^{-4} to $12 \times 10^{-4} \text{ m s}^{-1}$ if values at all locations (5 to 435 m downwind) are included in the comparison, Table 2. This comparison of deposition velocities is based on selection of soils having normal water contents (eg 13.5 to 42.0%, Table 1).

Comparison of Darlington HT deposition velocities with those of soils of similar water content at various locations indicates excellent agreement including the preliminary 1986 Chalk River experiment, Table 2. Most deposition velocities fell between 1×10^{-4} and $13 \times 10^{-4} \text{ m s}^{-1}$. These deposition velocities were measured both in the field and laboratory for a wide variety of soil textures, water contents (extremes excluded, eg <5%, >40% wt/dry wt) and soil biotype (field, forest and pasture). Exposure temperatures were those which normally support microbiological activity (above freezing). Field values included soils with and without vegetation cover.

TABLE 2: COMPARISON OF SOIL DEPOSITION VELOCITIES

Deposition Velocity (10^{-4} m s^{-1})	Water Content (% of dry wt)	Experiment	Location	Reference
2.7 to 12	4 to 39	Field	Chalk River	Brown <i>et al</i> 1988
4.1 to 13		Field	Chalk River	Brown <i>et al</i> 1987
0.7 to 7.2	6 to 12	Field	Wesleyville	Dunstall <i>et al</i> 1985
<0.05	frozen	Field	Wesleyville	Dunstall <i>et al</i> 1986
1.0 to 13	12 to 35	Lab ¹	3 Ontario soils ²	Spencer <i>et al</i> 1986
<0.2	25	Lab ¹	Wesleyville	Spencer <i>et al</i> 1986
0.7 to 3	3 to 53	Lab	11 Ontario soils	Dunstall <i>et al</i> 1985
0.2	295	Lab	Goodwood ³	Dunstall <i>et al</i> 1985
3.0	8 to 14	Field	German loess ⁴	Bunnenberg <i>et al</i> 1986
<0.6	<3, >17	Field	German loess ⁴	Bunnenberg <i>et al</i> 1986
0.9 to 5.5		Field	French soil ⁵	Belot <i>et al</i> 1986
7.1 to 8.2		Field	German soils	Diabaté <i>et al</i> 1986
15		Field	German soil ^{3,5}	Diabaté <i>et al</i> 1986
1.0 to 10		Lab	French soil	Förstel <i>et al</i> 1986
0.8 to 3.4		Field	South Carolina	Sweet and Murphy 1982
1.0 to 11		Field	UK soils	Garland and Sweet 1980

¹ Experimentally manipulated water content and porosity

² Mulliken sandy loam, Pickering silty-clay, and Wesleyville loamy sand

³ Value for a high organic marshy soil

⁴ A subsoil exposed during the 1986 French HT Release Experiment

⁵ Soils exposed during the 1986 French HT Release Experiment

The Chalk River HTO soil profile data from both the 1987 HT release and the preliminary 1986 release indicate that soil sampling to a depth of 22 cm included all HT deposited to soil as HTO (Brown *et al* 1987 and 1988). Most tritium, typically 80 to 95%, resides in the upper few cm following deposition.

Laboratory studies under controlled conditions have shown that deposition velocity depends on water content and soil porosity, parameters which are interrelated (Bunnenberg *et al* 1986 and Dunstall *et al* 1985). Deposition velocity usually rises by an order of magnitude within a normal range of decreasing water content and increasing porosity. The water content and porosity parameters may be expressed in various ways. In Figure 3 Darlington deposition velocities are plotted against free pore volume (air space not filled with water). The relationship appears to be reasonably linear over the range from about 5 to 35% free pore volume although with data from various soil textures an equation is not fitted. This provides a somewhat better fit than plots of deposition velocity against water content (data not shown). The deposition velocity at site 4, a bermed area, is clearly an outlier. The free pore volume is small because of the abnormally high bulk density value of 1.66 g cm^{-3} for a loam. This is consistent with the severe compaction noted during soil sampling and likely resulted from construction activity.

It is important to note that HT oxidation by soils would be severely curtailed by extremes of water content as shown in Table 2 by Bunnenberg *et al* (1986) for a German soil in the field and by Dunstall *et al* (1985) for Ontario soils in the laboratory. Seasonality in terms of temperatures near or below freezing is also an important variable, not as yet thoroughly investigated. At Wesleyville an upper limit deposition velocity of $0.05 \times 10^{-4} \text{ m s}^{-1}$ was measured over frozen soil, Table 2.

Deposition velocity is dependent on biological activity (reaction rate constant) and the rate of diffusion of HT into soil, expressed as the diffusion coefficient. This mechanistic relationship has been outlined in a report by Ogram (1984) and has been tested under laboratory conditions in a series of experiments by Dunstall in an Ontario Hydro Research Division report in preparation. Thereby either deposition velocity values or more directly diffusion coefficients and biological activity constants may be used in modeling HT oxidation to HTO in the environment.

CONCLUSIONS

A series of soil samples collected at Darlington and exposed to atmospheric HT during the 1987 Chalk River Field Experiment provided deposition velocities of $1.4 \times 10^{-4} \text{ m s}^{-1}$ to $11.6 \times 10^{-4} \text{ m s}^{-1}$. These values were in very close agreement with those measured during the preliminary 1986 HT release at Chalk River, the full-scale 1987 Chalk River HT release, and various other values obtained with exposure chambers under laboratory and field conditions.

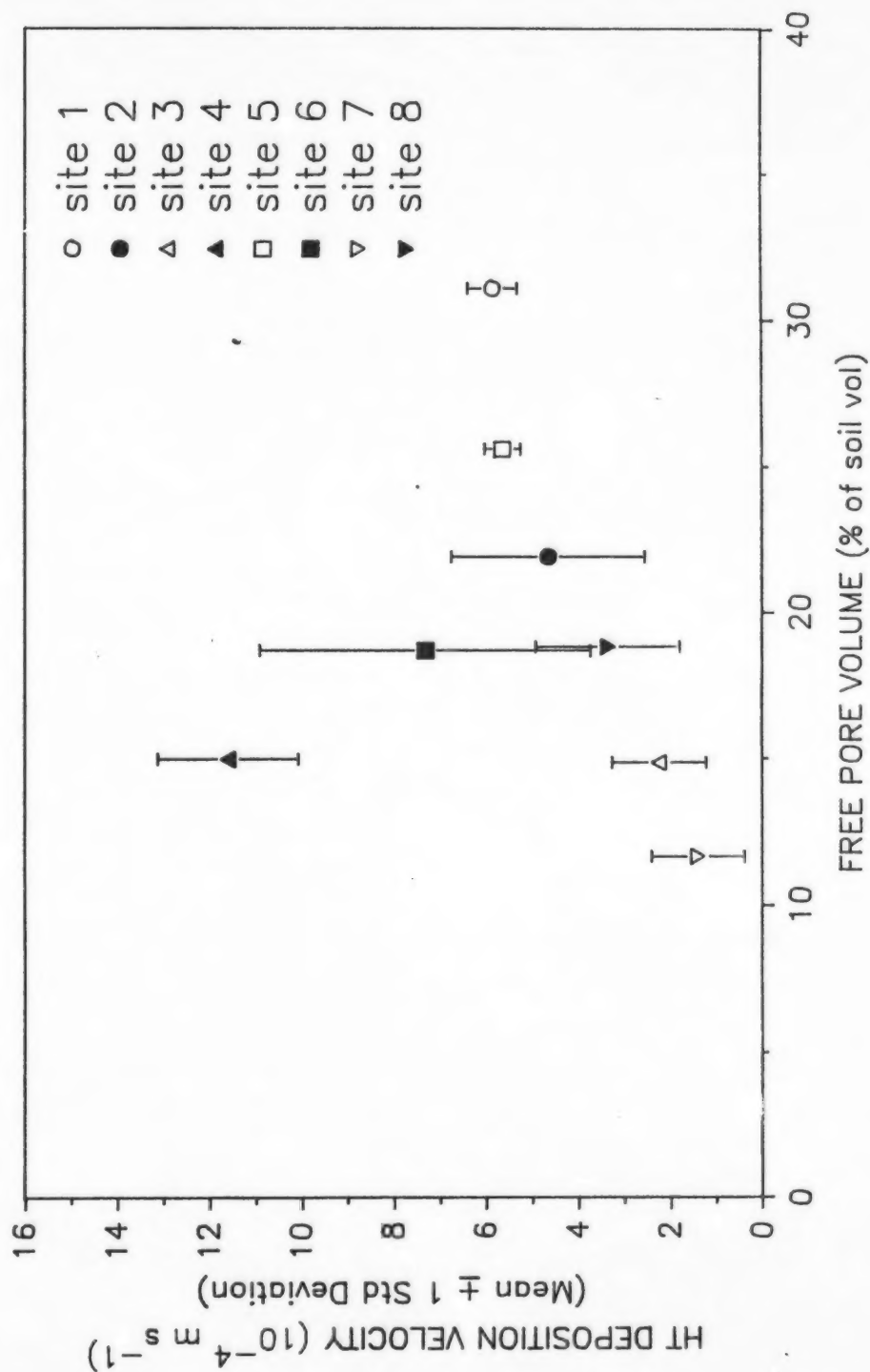
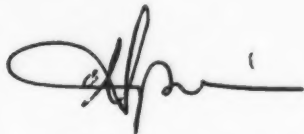


Figure 3. Deposition Velocity Dependence on Soil Free Pore Volume

It is concluded that the oxidation of HT released into the atmosphere at Darlington would be similar to that observed at Chalk River and at other sites. This is based on the similarity of soil HT deposition velocities and assumes a normal range of key soil parameters including water content and porosity and temperatures supportive of biological activity. It is expected that winter deposition velocities would be at least two orders of magnitude less than those observed above during summer.

Approved by:



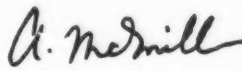
O.A. Kupcis
Manager
Chemical Research Department

Submitted by:



F.S. Spencer
Associate Research Scientist
Biological Research Section

Reviewed by:



A.C. McMillan
Section Head
Biological Research Section

FSS:cam:ra

REFERENCES

1. Belot, Y. Soil and Plant Measurements. In: Environmental Tritium Behaviour. French Experiment. Final Report Draft. H. Djerassi, and B. Lesigne (eds). Commissariat a l'Energie Atomique, IPSN, Issue 88/03 (1988).
2. Brady, N.C. The Nature and Properties of Soils. MacMillan Publishing Co, New York (1974).
3. Brown, R.M., Ogram, G.L. and Spencer, F.S. Field Studies of HT Behaviour in the Environment: I. The 1986 August Experiment at Chalk River, Canadian Fusion Fuels Technology Project Report No CFFTP-G-87004 (1987).
4. Brown, R.M., Ogram, G.L. and Spencer, F.S. Field Studies of HT Oxidation and Dispersion in the Environment. II. The 1987 June Experiment at Chalk River, Canadian Fusion Fuels Technology Project Report No CFFTP-G-88007 (1988).
5. Bunnenberg, C., Feinhals, J., and Wiener, B. Differences in the Behaviour of HTO and H_2O in Soil After Condensation from the Atmosphere and Conversion of HT to HTO and OBT in Soil Relative to Moisture Content and Pore Volume. Rad. Prot. Dosim., 16, 83-87 (1986).
6. Bunnenberg, C. Soil and Plant Measurements. In: Environmental Tritium Behaviour. French Experiment. Final Report Draft. H. Djerassi, and B. Lesigne (eds). Commissariat a l'Energie Atomique, IPSN, Issue 88/03 (1988).
7. Diabaté, S. Soil and Plant Measurements. In: Environmental Tritium Behaviour. French Experiment. Final Report Draft. H. Djerassi, and B. Lesigne (eds). Commissariat a l'Energie Atomique, IPSN, Issue 88/03 (1988).
8. Dunstall, T.G., Ogram, G.L. and Spencer, F.S. Elemental Tritium Deposition and Conversion in the Terrestrial Environment, Fusion Technol., 8, 2251-2256 (1985).
9. Förstel, H. Soil and Plant Measurements. In: Environmental Tritium Behaviour. French Experiment. Final Report Draft. H. Djerassi, and B. Lesigne (eds). Commissariat a l'Energie Atomique, IPSN, Issue 88/03 (1988).
10. Garland, J.A. and Cox, L.C. The Absorption of Tritium Gas by English Soils, Plants and the Sea. Water Air Soil Pollut., 13, 317 (1980).
11. Guenot, J. Soil and Plant Measurement. In: Environmental Tritium Behaviour. French Experiment. Final Report Draft. H. Djerassi, and B. Lesigne (eds). Commissariat a l'Energie Atomique, IPSN, Issue 88/03 (1988).
12. Förstel, H. Soil and Plant Measurements. In: Environmental Tritium Behaviour. French Experiment. Final Report Draft. H. Djerassi, and B. Lesigne (eds). Commissariat a l'Energie Atomique, IPSN, Issue 88/03 (1988).
13. Ogram, G.L. A Simple Model for the Evolution of the T_2 Profile in Soil Exposed to an Atmosphere Containing T_2 . Ontario Hydro Research Division Report No 84-260-K. (1984).
14. Russell, S.B. and Ogram, G.L. Modeling Elemental Tritium Deposition, Conversion and

Resuspension Using Ontario Hydro's Tritium Dispersion Code. Submitted to Fusion Technology (1988).

15. Spencer, F.S. and Dunstall, T.G. Molecular Tritium Conversion in Vegetation, Litter and Soil. Rad. Prot. Dosim., 16, 89-93 (1986).
16. Sweet, C.W. and Murphy, C.E., Jr. Oxidation of Molecular Tritium by Intact Soils. Environ. Sci. Technol., 15, 1485 (1981).

APPENDIX I

SOIL WATER CONTENT, HTO CONCENTRATION, HTO DEPOSIT AND HT DEPOSITION VELOCITY

HT Release Date: 10 June, 1987
 Release Duration: 1520 to 1550 h
 Release Site: AECL, Chalk River, Meteorological Field
 HT Air Concentration: $1.455 \times 10^6 \text{ Bq m}^{-3}$ (25 min avg at 50 m downwind)
 Samples Exposed: Darlington soil cores from at 8 sites (Fig 1)

Soil Site	Replicate No.	Sample Wet Wt (g)	Water Content (% by dry wt)	Sample Conc'n (Bq L ⁻¹)	Sample Activity (Bq)	Soil Deposit (Bq m ⁻²)	Vg x 10 ⁴ (m s ⁻¹)
1	1	916.3	21.0	27606	4391	1408770	5.38
1	2	952.2	25.6	27101	5263	1688605	6.45
1	3	1007.3	22.0	25871	4702	1508352	5.76
2	1	1010.2	37.8	7424	2056	659683	2.52
2	2	982.6	38.6	20125	5504	1765672	6.74
2	3	912.2	42.0	14295	3858	1237675	4.73
3	1	990.9	39.9	4211	1190	381866	1.46
3	2	1098.6	37.5	5079	1522	488196	1.86
3	3	1035.0	48.5	8243	2785	893418	3.41
4	1	1274.4	14.1	62722	9862	3163868	12.08
4	2	1296.5	12.4	73101	10479	3362040	12.84
4	3	1304.4	14.0	50361	8084	2593383	9.90
5	1	1039.4	29.1	20970	4908	1574544	6.01
5	2	954.0	24.1	25140	4661	1495284	5.71
5	3	1002.3	28.7	19116	4272	1370480	5.23
6	1	1055.4	27.4	27916	6341	2034451	7.77
6	2	1078.8	24.6	41003	8728	2799982	10.69
6	3	1119.9	32.6	10510	2892	927742	3.54
7	1	1198.8	26.9	8115	2064	662035	2.53
7	2	1309.5	17.5	4531	885	283934	1.08
7	3	1208.6	21.9	2173	472	151289	0.58
8	1	1146.2	23.5	6018	1311	420455	1.61
8	2	1119.1	21.9	18852	3787	1214975	4.64
8	3	1136.1	21.7	15537	3152	1011210	3.86